Ball Milling Simulation by Discrete Element Method and Its Application to Grinding and Mechanochemical Phenomena(離散要素法によるポールミルシミュレーションと粉砕ならびにメカノケミカル現象への適用)

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論文内容要旨

Grinding is one of the most important unit operation in many fields such as chemical, material, pharmaceutical, cement and mineral industries. The purpose of grinding is to be high reactivity with other phases, good liberability and easy handle performance. In addition, recycling of waste material is high interest in recent years, so that they are subjected to grind to make powder. It is well known that the grinding causes mechanochamical (MC) effects. This operation is able to dechlorinate halide compounds such as polyvinyl chloride and detoxificate PCB and dioxins. The grinding is operation which consumes a lot of energy, and the efficiency is quite low. Therefore, much effort has been attempted to improve it, but it has not been rewarded due to mostly empirical methods. Normally, the grinding efficiency depends on the ball motion, there are many reports on the analysis of ball motion, however, the most of them are not linked with the experimental results. It is still difficult to design a mill scaled-up and optimise the grinding operation.

In this Ph.D. thesis, the author has chosen typical two grinding mills; one is a tumbling ball mill and the other is a planetary mill. Fig.1 shows a snap shot of the ball motion in a tumbling mill, which has been used to grind materials in not only the laboratory but also the industry. Fig.2 shows a bird view of a planetary ball mill, in which the motion of balls is quite complicated. The author has attempted to simulate the ball motion in these mills by using the Discrete Element Method (DEM). The specific impact energy of balls during milling was proposed and calculated to find

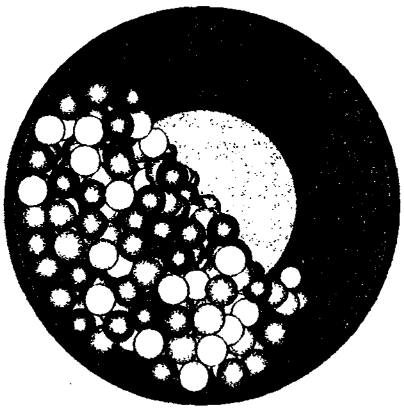

Figure 1 Ball motion in tumbling ball milling.

out the relationship to the grinding rate in the experiment. The impact energy is calculated from the relative velocity on collision at each time step, it is original and novel method for analysis of ball milling.

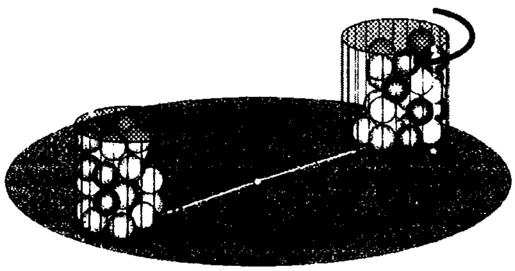

Figure 2 Ball motion in planetary ball milling.

Chapter 1 has surveyed the background of the grinding and the application of the ball milling to the Mechanochemical phenomena. The recent research of the analysis of ball milling by the simulation work was described. The general of DEM was given briefly.

Chapter 2 has introduced the tumbling ball milling simulation and the correlation between the grinding rate and the specific impact energy of balls. The balls' movement during milling was described exactly and the specific impact energy of balls was calculated under several grinding conditions. The grinding rate, K_P, of the gibbsite sample increased with increasing in the rotational speed of the mill, and it fell around the critical rotational speed, N_C, due to balls' rolling motion. It became large as the pot diameter increases when it is compared at the same N/N_C. K_P by using balls having 12.7 to 19.1mm of diameter dropped very quickly because of the relation between the ball diameter and its dead space when balls were in the rolling motion. However, K_P of smaller or larger balls than that range didn't drop so rapidly, because there is some possibility for the sample to be ground by few free balls or spaces between balls. The effect of pot length, l_M, on the grinding rate is less than that of pot diameter except for l_M=71mm. This pot is very narrow; therefore, there is large interactive force from side walls acting on balls. This force helps balls to be rolling state at the high rotational speed. K_P rapidly fell over N/N_C showing maximum value under the condition of not more than 30% of ball-filling ratio, while it slowly fell under not less than 40%. The rotational speed at the maximum value of K_P shifted to higher side as the ball-filling ratio decreases, and K_P became large. It can be seen the quite similar tendency between the specific impact energy, E_W, of balls by simulation and experimental results. Some simulated results showed the slightly different values, however, the most of data had a well comparison. K_P of the most of grinding conditions were correlated with the specific impact energy, and its relationship can be expressed by a straight line. Thus, the grinding rate in a tumbling ball milling under any conditions will be estimated by this relationship.

Chapter 3 has introduced the planetary ball milling simulation and the correlation between the grinding rate and the specific impact energy of balls. Four kinds of materials were ground in the experimental work and the grinding rate of each sample was examined. The grinding rate increased with an increment of the revolution speed of the planetary ball mill. It was given large value when the small amount of powder sample was loaded, the heavy balls were put in a pot or the large volume of pot was introduced in the mill. The mixed balls work well because small balls can be in the dead space between large balls. The similar tendency as grinding rate was seen in the simulated results under all conditions. The grinding rate is proportional to the specific impact energy very well, and this relationship can be expressed by a liner equation. The prediction of the grinding rate was tried by using this relationship. The predicted size reduction curves were well correlated with the actual size reduction. The usage of the specific impact energy for the prediction of the grinding performance was proved, and the specific impact energy would be a key factor in the grinding process. The effect of the ground materials was observed by the difference of the gradient of the relationship between K_P and E_W. Values of gradients in the straight lines for all samples were inversely proportional to a net work index, W_{in}, which was calculated by substituting $E_W \times t$ instead of W_P in the Bond's third theory. Therefore, K_P for other materials can be estimated by using this work index.

Chapter 4 has proposed the usage of the ball mill simulation and the impact energy for the industrial applications. Four kinds of liner design (MODEL I, II, III and IV) in a tube mill were modelled by the computational simulation to evaluate the effect of liner design on the grinding performance. MODEL I had the wavy liner, the liner of MODEL II was like steps. MODEL III had the wavy liner on the steps. MODEL IV was one of most traditional designs, which likes a trapezoidal shape, and MODEL O has no liner inside mill. The Balls' movement of MODEL I, III and IV were cataracting motion, and movement in MODEL II seemed to be cascading and cataracting motions. However, the cascading motion was seen in MODEL O in spite of the same rotational speed. Thus, the liner can raise balls to the higher position. MODEL I, III and IV had the larger impact energy than that of MODEL O, it was found that the wavy and trapezoidal liners work well for the grinding. MODEL III had the largest normal impact energy in all mills; it is improved 32% than non-liner mill. The existence of the liner in a tube mill can make a collision having the large impact energy, it can be observed from the impact energy distribution curves. There was huge difference between distribution curves of tube mills with liner and a curve of non-liner mill. The liner works for the raising balls and balls hit on the bottom mill wall directly, which was observed by the mapping of the impact energy fields. The hot area of the impact energy in MODEL O and II are the middle of the balls' flow surface due to the cascading motion. Therefore, the existence of the liner in a tube mill is worthy for the grinding, especially design of MODEL III works well. This is the useful information for the design of the tube mill and determination of the grinding condition.

Chapter 5 proposed the guideline for the scale-up and optimisation of ball mills by the impact energy. The impact energy, E_i, in a tumbling ball mill was proportional to the pot length because of an increment of the number of balls, n_B, charged into the pot. The proportional constant in this relationship depended on the ball-filling ratio strongly when the pot diameter, d_M, was constant. E_i depended on d_M^{λ}, whose index number, λ, showed the range from 2.25 to 2.6, which are very similar to past-research. λ was divided as λ_{n_B} and λ_D. λ_{n_B} was the index number obtained from the effect of the number of balls, and λ_D was from the square of balls' maximum relative height. The proportional constant in this relationship of the effect of the pot diameter also depended on the ball-filling ratio. E_i increased with increasing the ball-filling ratio, and it decreased over 55% of filling ratio, which seemed the optimum condition under all pots. The relation between E_i and J can be expressed by the cubic equation. The impact energy of balls in the planetary ball mill was proportional to d_M^3 due to the increment of number of balls and the lengthened way for the acceleration by an increment of d_M. The linear relationship was seen against the pot depth, h, and the revolution radius, R. The relationship between the impact energy and the total scale-up ratio was dependent on the fifth power of the scale-up ratio. The rotation in the counter direction in a planetary ball mill can improve the impact energy of balls rather than that in the normal direction. This result was confirmed in the experimental work of the grinding talc. The impact energy of balls increased with an increasing in the rotation-to-revolution speed ratio, however it falls over about the critical speed ratio due to rolling motion. The critical speed ratio can be determined by the simple equation, which depends on the revolution radius and the pot diameter. It is important to keep the milling condition at the critical speed ratio for effective milling.

Chapter 6 has investigated the application of the ball mill simulation to the mechanochemical (MC) phenomena. The dechlorination of polyvinyl chloride (PVC) and the extraction of the yttrium from the fluorescent powder were carried out to find out the relationship between the MC reaction rate and the specific impact energy, and the optimisation of a planetary ball mill for the MC reactor was discussed. MC dechlorination of PVC proceeded with the grinding progress, and it was improved with increasing in both the revolution speed of a mill and the amount of balls charged into the pot. The specific normal impact energy, E_N, can express the value of dechlorination rate in the experiment very well. The relation between the dechlorination rate and the specific normal impact energy was given by a straight line regardless of the MC conditions. Therefore, the dechlorination rate of PVC is proportional to the specific normal impact energy of balls calculated from the computer simulation. This relation of the small mill can be applicable to the treatment in the large mill regardless of the ball diameter. The dechlorinating curves predicted by E_N were correlated with the actual data from the experiment much better than those predicted by E_W. The extraction yield of yttrium (Y) proceeded with milling progresses, and the extraction rate was improved with an increase in the revolution speed. However, it was independent on the ball diameter put into a pot. The extraction rate of Y was proportional to the specific normal impact energy of balls calculated from the computer simulation, irrespective of the milling conditions. The relation between the extraction rate and the specific normal impact energy for the small-scale mill was applicable for predicting the extraction rate using the large-scale mill. This indicates that the extraction yield and its rate of Y for any large mill can be predicted by substituting the specific normal impact energy of the balls into the relation between K_e and E_N, obtained from the small-scale mill. These correlations have already seen in the dechlorinating process of PVC. Therefore, the specific normal impact energy is a key factor to dominate the mechanochemical phenomena in ball milling. It is found from the simulation work that the normal impact energy was independent on the ball diameter. This have already provided in the experimental work of extraction of yttrium. The grinding condition of the ball-filling ratio is most important things for mechanochemical reaction in the planetary ball mill. The best ball-filling ratio is obtained at 60 to 65 % of pot volume.

Chapter 7 is composed of summaries about all chapters, as follows; the simulation of the ball milling has been conducted by DEM. The ball motion was described exactly same as the actual motion in tumbling and planetary ball mills, and the correlation between the grinding rate and the specific impact energy of balls has been shown. The guideline of the mill design and the scale-up method of the ball mill have been proposed by the impact energy, and the liner design in a industry scale tube mill have been also discussed. The dominant factor of the mechanochemical (MC) phenomena has been found out by the simulation; the MC reaction is depends on the normal component of the impact energy. This investigation would be very useful to scale-up of MC treatment in the industry field.

論文審査結果の要旨

粉砕はエネルギー効率の極めて低い操作である。それにもかかわらず、この操作はセメント製造工場等をはじめとする多くの物質生産工業分野で取り入れられ、そこでの消費エネルギーの総量は500億KWhにも達するといわれる。粉砕のエネルギー効率向上は、省エネルギー・省資源に大きく寄与し、操作条件やミル構造の最適化が望まれる。それには、これまでのような実験データに基づく経験則では無い、理論的・普遍的な最適操作条件や効果的なミル構造の決定法を提案することが重要になっている。

本論文は、ミル操作条件や構造の最適化を行う上で必要な媒体運動を的確に再現できるシミュレーション法を提案し、得られる情報が粉砕やメカノケミカル現象を高精度で予測でき、また、ミルのスケールアップに対しても十分活用できることを明確にした研究内容であり、全編6章よりなる。

第1章は序論である。

第2章は、ミル内の一部、側面近傍でのボール運動の可視化が可能な転動ミルを対象とし、まず、水酸化アルミニウム（ギブサイト）試料共存下での粉砕におけるボール運動をシミュレーションし、その結果が可視化実験での結果と良好に一致し、シミュレーションにおいて決定したモデルとパラメータの妥当性を示した。その上で、粉砕速度定数の最大値を示すミル回転速度でのシミュレーション法から求められるボール運動エネルギーと粉砕速度定数とが良好に相関することを確認し、この結果はミル径とミル奥行き、ボール径、ボール充填率を変化させても同様な相関性を示すことを明確にした。このことは、本シミュレーション法が適切であり、したがって、転動ミル内での粉砕現象が、最低限度1回の実験を行うことによって、的確に予測可能であることを明確にした。

第3章は、ミル内でのボール運動の可視化が困難な遊星ミルを対象とし、転動ミルとは境界条件が異なるボール運動のシミュレーション法を提示し、ギブサイト試料のポット充填量、ボール密度、ボール径分布、ミルスケールを変化させた実験から得られる粉砕速度定数と本シミュレーション法から得られるボール運動エネルギーとの関連性を検討している。ここでも、最低限1回の粉砕実験を行い、これとシミュレーション法から得られるボール運動エネルギーとの相関性が良好に表示でき、したがって、任意の操作条件、ミルスケールでの粉砕における粉砕速度定数が予測できることを確認している。

第4章は、前章までの知見を踏まえ、セメント工業で活用されている内径 3.5m の大型チューブミル（ボール径が 60～90mm）に適用し、本シミュレーション法が十分活用できることを明確にしている。特に、ボール運動はミルライナー形状によって変化するが、チューブミルに採用されている 4 種類のミルライナーの形状をモデル化し、ボール運動を再現しているが、その結果は、現場での粉砕結果とライナー磨耗状況などを十分説明するに足る有用な情報を提供しており、本シミュレーション法の信頼性が確認されている。

第5章は、転動ミルならびに遊星ミルを対象とし、ボール運動のシミュレーション法に基づき、ミルの直径や長さなどの寸法を幾何学的相似でスケールアップさせた場合の最適操作条件決定法について述べている。すなわち、粉砕現象を支配するボール運動エネルギーを基準した場合、スケールアップ比が適宜決定できるなど、有用な情報を多く含んでいることが示されている。

第6章は、本シミュレーション法により得られるボール運動エネルギーが、遊星ミル内で起こるメカノケミカル現象を支配する主要な因子であることを明確に示した内容である。

第7章は結論である。

以上、要するに本論文は、離散要素法によるミル内ボール運動を正確に表示する手法を提案し、それを粉砕ならびにメカノケミカル現象へ適用し、その妥当性を明確にしたものであり、地球工学、素材工学の発展に寄与するところ少なくない。

よって、本論文は博士（工学）の学位論文として合格と認める。